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TECHNICAL FIELD

[Industrial Application] This invention relates to the hydrogen generating approach of having used electrochemistry equipment.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Industrial Application] This invention relates to the hydrogen generating approach of having used electrochemistry equipment.
[0002]
[Description of the Prior Art] Conventionally, there is a reformer as equipment for generating hydrogen from a hydrocarbon fuel.
[0003] There are a steam reforming process which a steam is added [steam reforming process] to material gas and transforms coal-for-coke-making-ized hydrogen to hydrogen, a carbon monoxide, and a carbon dioxide, and the partial oxidation method (partial combustion process) for burning a part of coal-for-coke-making-ized hydrogen and obtaining hydrogen, a carbon monoxide, and a carbon dioxide in the hydrogen generating approach performed in a reformer.
[0004] Generally it is Cn Hm about a hydrocarbon. If expressed, the reaction which occurs the reaction which occurs in a steam reforming process in [-izing 1] and a partial oxidation method can be shown in [-izing 2], respectively.
[0005]
[Formula 1]

$$C_nH_m + nH_2O \rightarrow nCO + (n + m/2)H_2$$

[0006]
[Formula 2]

$$C_nH_m + nO_2 \rightarrow nCO + m/2\,H_2$$

[0007] In these reactions, liftings and these reactions maintain a balance for the reaction which shows some carbon monoxides below, and it goes on.
[0008]
[Formula 3]

$$CO + H_2O \rightleftharpoons CO_2 + H_2$$

[0009]
[Formula 4]

$$CO + 3H_2 \rightleftharpoons CH_4 + H_2O$$

[0010] [-izing 1] is endothermic reaction and other reactions are exothermic reaction. Therefore, it is necessary to heat from the outside and a reaction is usually performed in the temperature of 800 degrees C - 900 degrees C by the coil of heat-resistant metal filled up with the catalyst of a nickel system at the reaction of [-izing 1]. On the other hand at such temperature, the reaction of [-izing 3] and [-izing 4] seldom advances on right-hand side, for example, in the case of a 850-degree C

reaction, the concentration of methane of the concentration of a carbon dioxide is 3 – 4% 13 to 14%.

[0011] Moreover, when the methanol which is a kind of alcohol instead of a hydrocarbon is used, reforming advances at a reaction like water and a degree type.

[0012]

[Formula 5]

$$CH_3OH + nH_2O \longrightarrow (1-n)CO + nH_2O + nCO_2 + (2+n)H_2$$

$$(0 < n < 1)$$

[0013] This reaction is 0–20kg/cm2. It is made desirable to set it as a pressure and the temperature of 200–600 degrees C.

[0014]

[Problem(s) to be Solved by the Invention] After performing the reaction mentioned above, the carbon monoxide of remarkable concentration is contained in the gas which comes out of a reformer.

[0015] For this reason, if gas leakage should occur, the risk of carbon monoxide poisoning follows. Moreover, in case the gas which comes out of a reformer is used for catalytic reaction, the carbon monoxide contained in this gas may carry out poisoning of the catalyst, and may bar a normal reaction.

[0016] Then, equipment called a carbon monoxide converter is used in order to make the amount of survival of a carbon monoxide low, and to push the reaction of [–izing 3] on the right. In order to push the reaction of the [–izing 3] which is exothermic reaction, reaction temperature is first set as 350 degrees C – about 370 degrees C, in order to raise a reaction rate, the so–called elevated–temperature inversion (Hot Shift) is performed using catalysts, such as an iron–chrome system, and low–temperature inversion (Cold Shift) is made to perform at about 200–230 degrees C further with a carbon monoxide converter using the catalyst of a copper–zinc system. Thereby, carbon monoxide concentration can be decreased.

[0017] When a fuel is alcohol, in order to reduce CO of reformed gas further, CO shift catalyst is used after this reforming reaction. In CO shift processing, CO is reduced by even about 1% of concentration by the reaction with H2 O. Furthermore, in order to reduce CO concentration, it can decrease to 100 ppm by making reformed gas react with air further with 2nd CO reduction equipment. However, by such approach, equipment becomes complicated and there is a problem of requiring an elevated temperature etc.

[0018] The purpose of this invention is to offer the approach of generating hydrogen gas from a fuel, without generating most carbon monoxides not using the above complicated flow systems.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] After performing the reaction mentioned above, the carbon monoxide of remarkable concentration is contained in the gas which comes out of a reformer.
[0015] For this reason, if gas leakage should occur, the risk of carbon monoxide poisoning follows. Moreover, in case the gas which comes out of a reformer is used for catalytic reaction, the carbon monoxide contained in this gas may carry out poisoning of the catalyst, and may bar a normal reaction.
[0016] Then, equipment called a carbon monoxide converter is used in order to make the amount of survival of a carbon monoxide low, and to push the reaction of [-izing 3] on the right. In order to push the reaction of the [-izing 3] which is exothermic reaction, reaction temperature is first set as 350 degrees C - about 370 degrees C, in order to raise a reaction rate, the so-called elevated-temperature inversion (Hot Shift) is performed using catalysts, such as an iron-chrome system, and low-temperature inversion (Cold Shift) is made to perform at about 200-230 degrees C further with a carbon monoxide converter using the catalyst of a copper-zinc system. Thereby, carbon monoxide concentration can be decreased.
[0017] When a fuel is alcohol, in order to reduce CO of reformed gas further, CO shift catalyst is used after this reforming reaction. In CO shift processing, CO is reduced by even about 1% of concentration by the reaction with H2 O. Furthermore, in order to reduce CO concentration, it can decrease to 100 ppm by making reformed gas react with air further with 2nd CO reduction equipment. However, by such approach, equipment becomes complicated and there is a problem of requiring an elevated temperature etc.
[0018] The purpose of this invention is to offer the approach of generating hydrogen gas from a fuel, without generating most carbon monoxides not using the above complicated flow systems.
[0019] The further purpose of this invention is from the fuel containing a methanol and water to offer the approach of generating hydrogen, without generating most carbon monoxides electrochemically.

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MEANS

[Means for Solving the Problem] The electrochemistry equipment used for this invention is formed in one pair of front faces on which the cation exchange membrane which has one pair of front faces which counter, and cation exchange membrane counter, respectively, and is equipped with the fuel-supply means for supplying a fuel to one side of one pair of electrodes which consist of ingredients including a catalyst, and one pair of electrodes. With the electrochemistry equipment used for this invention, the cation which is made to generate a cation and by which while a fuel is supplied was generated from the fuel in the electrode is transformed to a molecule by electronic supply on one pair of another side of an electrode through cation exchange membrane.
[0021] In this invention, cation exchange membrane will not be limited, especially if a cation is made to penetrate alternatively. As cation exchange membrane, the solid-state polyelectrolyte film, the film which consists of a matrix containing a phosphoric acid, the film which consists of a matrix containing a sulfuric acid, the film which consists of a solid electrolyte can be mentioned. Moreover, the ion exchange membrane which has a sulfonic acid, phosphonic acid, a sulfate, phosphoric ester, etc. can be used as a cation-exchange radical.
[0022] In this invention, the alloy of platinum, palladium, a rhodium, a ruthenium, gold, iridium, and these elements which contains either at least can be used for the catalyst contained in an electrode.
[0023] One pair of electrodes may be formed from the catalyst ingredient itself, such as an alloy of platinum, palladium, a rhodium, a ruthenium, gold, iridium, and these elements which contains either at least, and may be formed from the thing which made the conductive carbon electrode etc. support a catalyst ingredient.
[0024] The electrode according to this invention can be formed by depositing an electrode material in the porous structure on the front face of the film of cation exchange membrane. In addition, the electrode according to this invention can form a catalyst ingredient in a conductive electrode substrate electrolysis, by carrying out electroless deposition, etc.
[0025] In the equipment used for this invention, since a fuel is contacted to one side of an electrode, it can have a pump for sending a fuel into the container or tub which holds a fuel, this container, or a tub by the predetermined pressure etc., but a fuel-supply means is not limited to these, but if it is a device which supplies a fuel to an electrode, what kind of thing can also be used for it.
[0026] A fuel can be made to generate a hydrogen ion and the electrode of another side to a hydrogen content child from one electrode in the equipment used for this invention using what contains a methanol and water at least. In such equipment, the alloy of platinum, palladium, a rhodium, a ruthenium, gold, iridium, and these elements which contains either at least is preferably used as a catalyst included in an electrode.
[0027] Moreover, in the equipment made to generate the hydrogen mentioned above, the film which consists of an ingredient which can make hydrogen penetrate alternatively to the electrode side of

another side can be prepared. Such film can be named as a hydrogen permeable film, and its permeability of a hydrogen content child is high compared with the permeability of other matter. The film which consists of hydrogen storing metal alloys, such as the film which consists of macromolecules, such as polyimide and polystyrene, film which consists of a Pd alloy, Ti system, Mm system (Mm shows a misch metal), La system, and Mg system, etc. as such film can be used.
[0028] The approach of generating hydrogen with the equipment mentioned above can be offered. Moreover, this approach To an electrode including the catalyst which prepared one pair of electrodes in both sides to which cation exchange membrane counters, and was prepared in one side The reaction which generates a hydrogen ion is advanced from a methanol and water on an electrode by contacting the fuel which contains a methanol and water at least, impressing an electrical potential difference to one pair of electrodes, and picking out an electron from an electrode. It is characterized by changing the generated hydrogen ion into a hydrogen content child by electronic supply in the electrode prepared in another side of one pair of fields where cation exchange membrane counters.

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OPERATION

[Function] The mechanism of action of this invention is explained below using drawing.
[0030] Drawing 1 is the mimetic diagram showing an example of electrochemistry equipment according to this invention. In electrochemistry equipment 100, the electrode 102 for fuels and counterelectrode 103 which made the platinum catalyst support, respectively are prepared in the both sides of the cation exchange membrane 101 which consists of solid-state polyelectrolyte film.
[0031] The zygote which consists of such cation exchange membrane and one pair of electrodes can be inserted among separators 106 and 107. In the separator by the side of the electrode 102 for fuels, it is -cum- CO2 for fuel supply. Slot 108 a-e for discharge is formed further, and slot 109 a-e for hydrogen discharge is further formed in the separator 107 by the side of a counterelectrode 103.

[0032] In the electrode 102 for fuels, it is -cum- CO2 for fuel supply in the condition of a liquid or a gas about a fuel. By passing slot 108 a-e for discharge, a fuel is supplied and it is contacted by the electrode.
[0033] The electrode 102 for fuels and a counterelectrode 103 are electrically connected to an external circuit 104 respectively through separators 106 and 107. Between two electrodes, an electrical potential difference is impressed and a plus and counterelectrode 103 side is considered for the electrode 102 side for fuels as minus.
[0034] In the equipment mentioned above, water or a steam is supplied to the electrode 102 for fuels with the methanol which is a fuel, and through an external circuit 104, an electrical potential difference is impressed so that an electron may be drawn out from the electrode 102 for fuels. Consequently, the next reaction advances in the electrode 102 for fuels.
[0035]
[Formula 6] CH3OH+2H2 O->CO2+6e-+6H+[0036] Thus, the generated hydrogen ion passes cation exchange membrane, and is changed as follows with a counterelectrode 103.
[0037]
[Formula 7] 6H++6e-->3H2[0038] Such a process can be made to generate hydrogen alternatively by the counterelectrode 103 side. Therefore, according to the process of this invention, generation of CO is controlled.
[0039] Moreover, if the gas produced with the counterelectrode is collected through the hydrogen permeable film mentioned above, the concentration of a steam or other impurities can be reduced and hydrogen gas with high purity can be obtained.

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EXAMPLE

[Example]
Hydrogen gas was manufactured using the equipment shown in example 1 drawing 1 .
[0041] In this example, the electrode was formed as follows. NAFION117R (Du Pont make) was used as a cation exchange membrane which consists of solid-state polyelectrolyte film. As a metal salt, it is 1%NaBH4 as 3% chloroplatinic acid liquid and a reducing agent. After making a reducing agent permeate cation exchange membrane using a solution, the film front face was contacted in the chloroplatinic acid solution, and the platinum layer was deposited. By this approach, the porous platinum catalyst layer was formed in membranous both sides, and it considered as one pair of electrodes.
[0042] Thus, in the equipment of drawing 1 incorporating the zygote with which the electrode was formed in membranous both sides, the mole ratio supplied the methanol-water mixed liquor of methanol:water =1:2, and set the temperature of the cel which consists of a zygote as 30 degrees C at the electrode 102 side for fuels. And when the electrode 102 side for fuels was considered as plus and the electrical potential difference of 0.2V was applied between the electrode 102 for fuels, and the counterelectrode 103, it is 1.0 mA/cm2. A current flows and it is CO2 from the electrode 102 for fuels. Hydrogen gas was generated from the counterelectrode 103. The carbon monoxide was not detected in the generated hydrogen.
[0043] In the equipment of example 2 example 1, the polyimide poly membrane was prepared as a hydrogen permeable film on the counterelectrode, and hydrogen was collected through this film. When the electrical potential difference of 0.2V was impressed between the electrode for fuels, and the counterelectrode on the same conditions as an example 1, it is 1.0 mA/cm2. A current flows and it is CO2 from the electrode for fuels. Hydrogen gas was generated from the counterelectrode. The carbon monoxide was not detected in the hydrogen collected through the polyimide poly membrane.
[0044]

[Translation done.]

EFFECT OF THE INVENTION

[Effect of the Invention] According to this invention, the complicated system of reaction and the complicated flow system like before cannot be required, and can make very simple equipment generate hydrogen with high purity, as explained above. According to this invention, generation of impurities, such as CO, is controlled.
[0045] For this reason, while the risk of the poisoning of carbon mono oxide is avoided, the fall of the chemical reactivity by poisoning of a catalyst is also prevented about the hydrogen gas generated.

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CLAIMS

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(57) [Claim(s)]
[Claim 1] To an electrode including the catalyst which prepared one pair of electrodes in both sides to which cation exchange membrane counters, and was prepared in one side The reaction which generates a hydrogen ion is advanced from said methanol and water on said electrode by contacting the fuel which contains a methanol and water at least, impressing an electrical potential difference to said one pair of electrodes, and picking out an electron from said electrode. The hydrogen generating approach characterized by changing said generated hydrogen ion into a hydrogen content child by electronic supply in the electrode prepared in another side of one pair of fields where said cation exchange membrane counters.

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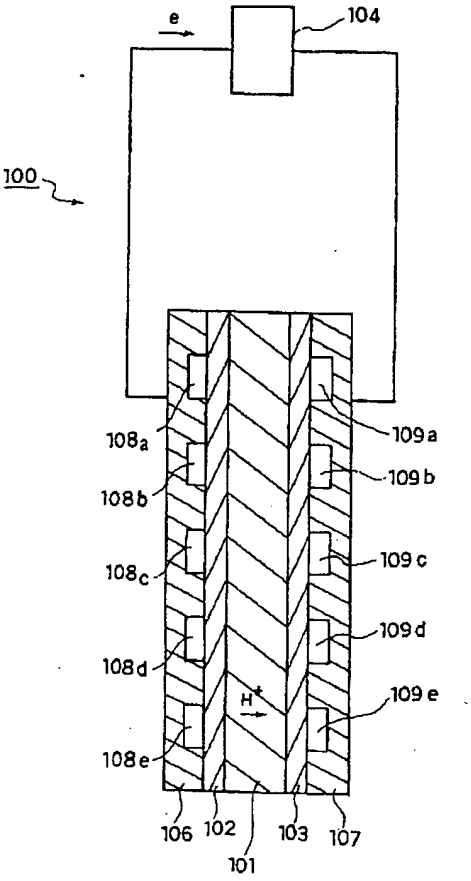
DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] It is the outline sectional view showing one example of electrochemistry equipment of following this invention.
[Description of Notations]
100 Electrochemistry Equipment
101 Cation Exchange Membrane
102 Electrode for Fuels
103 Counterelectrode
104 External Circuit
106 Separator
107 Separator

[Translation done.]

Drawing selection drawing 1



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